DENDRITIC GROWTH VELOCITY OF ICE CRYSTALS

K. OHSAKA* and E. H. TRINH

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109, USA

Abstract

The dendritic growth velocity of ice crystals is measured at undercooking levels between 5 - 8.5 K using a unique apparatus. The result shows that the measured velocities are smaller than the theoretical prediction based on the thermal diffusion theory with the scaling constant, $\sigma = 0.025$, which has been shown to agree with the experimental results at lower undercooking levels between 0.4 - 2.5 K. This deviation may be attributed to sluggish interface kinetics which start playing a significant role in dendritic growth at these undercooking levels.

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Keywords: Dendritic ice; Growth velocity; Undercooking; Interface kinetics

* Corresponding author:
Address: Jet Propulsion Laboratory
California Institute of Technology
MS 183-401
4800 Oak Grove Drive
Pasadena, CA 91109-8099
USA
Telephone: +1 8183543111
Fax: +1 8183935039

Dendritic crystal growth is a common growth pattern observed in many metals, alloys and other materials when they are solidified under typical solidification conditions. The evolution of the dendritic pattern has received much attention from both scientific and engineering points of view for its intricate pattern selection mechanisms and useful industrial applications. Succinonitrile and water are the most thoroughly studied materials among many materials which show dendritic growth. Succinonitrile is an ideal model material in terms of transparency, shape isotropy and solidification temperature; therefore, it is widely used to test the theoretical predictions of dendritic growth [1]. On the other hand, water is highly anistropic and complicated, showing the morphological change as it grows, but it is the most common and important material [2-5]. The morphology of ice crystals changes from a circular disk, to a perturbed disk and then to a dendrite as the ice grows [5]. The growth velocity of dendritic ice at undercooking levels below 4.5 K has been measured and compared with the theoretical predictions [5). This comparison shows that the data agree with the prediction at undercooking levels between 0.4 -2.5 K, but start deviating from theoretical prediction at undercooking levels below 0.4 K due to convection in the undercooked water. The data above 2.5 K also show some deviation from the prediction. The growth velocity at higher undercooking levels above 4.5 K will be useful to better pinpoint the upper limit of the theoretical prediction.

We have developed a unique apparatus whose primary purpose is to demonstrate dynamic nucleation of ice induced by cavitation in undercooked water [6], but it can also be used to observe the free growth of ice crystals. Figure 1 shows the schematic diagram of the apparatus. The levitation cell is filled with a water/ethylene glycol mixture in which a stationary acoustic wave is excited at around 21 kHz by the transducer. The cavitation cell is made from a thin wall plastic tube closed at the bottom, and it is filled with distilled water (approximately 3 ml) which has been passed through a 5 μm filter. The cooling of the cells is accomplished by circulating chilled fluid through the copper tubing. The mixture temperature is monitored by a thermocouple placed close to the cavitation cell. The

experimental procedure involves trapping of a micron-size bubble in the undercooked water by an ultrasonic field which also drives the bubble in a large amplitude volume oscillation mode (cavitation mode). When the conditions are satisfied, ice is nucleated near the center of the cavitation bubble because of the melting point shift caused by a high pressure pulse associated with the collapsing bubble. Once ice is nucleated, it freely grows into the undercooked water. It should be emphasized that the dendrite in the present apparatus freely grows without arty support by a substrate. In previous studies [3-5], although the core of ice was actually anchored by a capillary tube, the advance of the dendritic tips was described as free growth. The sequence of nucleation and growth of ice is monitored by a video camera and recorded on video tapes for post experiment analyses. In this letter, we report the growth velocity of ice determined by measuring the advancing dendritic tips recorded as the video images.

Figure 2 is a sequence of the images which show the growing dendritic ice in the undercooked water whose undercooking level is 5 K. The time interval between the consecutive images is 1/30 second. After nucleation, it appears that the growth of ice follows the morphological change described above; however, the pre-dendritic patterns are not visible due to their small sizes and short life times. The dendritic tips generally advance along the basal plane, (0001). The overall hexagonal shape is preserved at this stage. The growth velocity is determined by measuring the distance from the center of the crystal to the advancing tips. For the actual velocity determination, only the tips which are advancing on the plane perpendicular to the observation direction are selected. For the dendrite in Fig.2, those are growing upwards and downwards.

Figure 3 shows the result of the velocity measurement. We could measure the velocities at undercooking levels between 5 - 8.5 K. The lower limit of 5 K is the experimental threshold for cavitation bubble induced nucleation. The upper limit of 8.5 K is due to the rapid nucleation rate which prevents further undercooking. The data indicated by the squares are a partial reproduction of Langer et al.'s data [3]. The experimental results are

compared with the theoretical prediction based on the thermal diffusion model [7]. The solid line is the theoretical prediction with the scaling constant, $\sigma = 0.025$ which has been shown to agree well with the experimental data at undercooking levels between 0.4 -2.5 K [5]. The theoretical prediction runs between Langer et al's data and the present data. Due to the limited magnification and spatial resolution of current video imaging, the tip radius could not be determined for an independent check of the scaling constant.

We have utilized the unique apparatus which can nucleate a single crystal of ice in the undercooked water to observe the subsequent growth of ice. The observation allows us to determine the dendritic growth velocity of ice at undercooking levels between 5.0 -8.5 K. The previous maximum undercooking level is approximately 4.5 K [3]. The technique used to measure the velocity is simple and thus prone to various errors. For example, we had no control over the growth direction of the tips; therefore, we had to select the tips which happened to grow on the plane perpendicular to the observation direction. The selection is somewhat subjective and may introduce significant error. The true water temperature may be off by at most 0.3 K because of the indirect measurement. The thermocouple could not be immersed in the undercooked water because it would initiate nucleation. To estimate the magnitude of the error, the repeated measurements (5 times) were performed at the undercooking level, 5 K. As seen in Fig. 3, the data varies approximately $\pm$ 10 % from the average value. It is likely that scattering increases as the undercooking increases because of the increasing growth velocity. However, these errors cannot totally account for the discrepancy between the present data and the theoretical prediction. The departure of the present data from the theoretical prediction is opposite from that of Langer et al.'s data. The them-ml diffusion theory assumes local thermal equilibrium at the growing interface. However, this may not be valid at higher undercooking levels if the water molecule transfer across the interface is slow. Sluggish interface kinetics results in a slower growth velocity

than the theoretical prediction [8]. The direction of the departure of the present data from the theory is consistent with this interpretation.

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Captions

Figure 1. Schematic diagram of the experimental apparatus.

Figure 2, Sequence of the dendritic growth of the ice crystals. The interval between the consecutive images is 1/30 second.

Figure 3. The dendritic growth velocity of ice crystals as a function of undercooking. The data indicated by the squares area partial reproduction of Langer et al.'s data [3]. The solid line is the theoretical prediction based on the thermal diffusion theory with the scaling constant, $\sigma = 0.025$ [5].

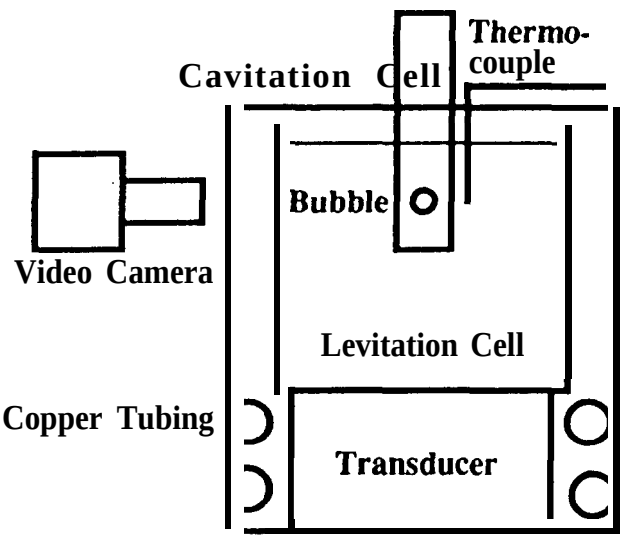
Thermo-
couple
Cavitation Cell
Bubble
Video Camera
Levitation Cell
Copper Tubing
Transducer

Figure 1

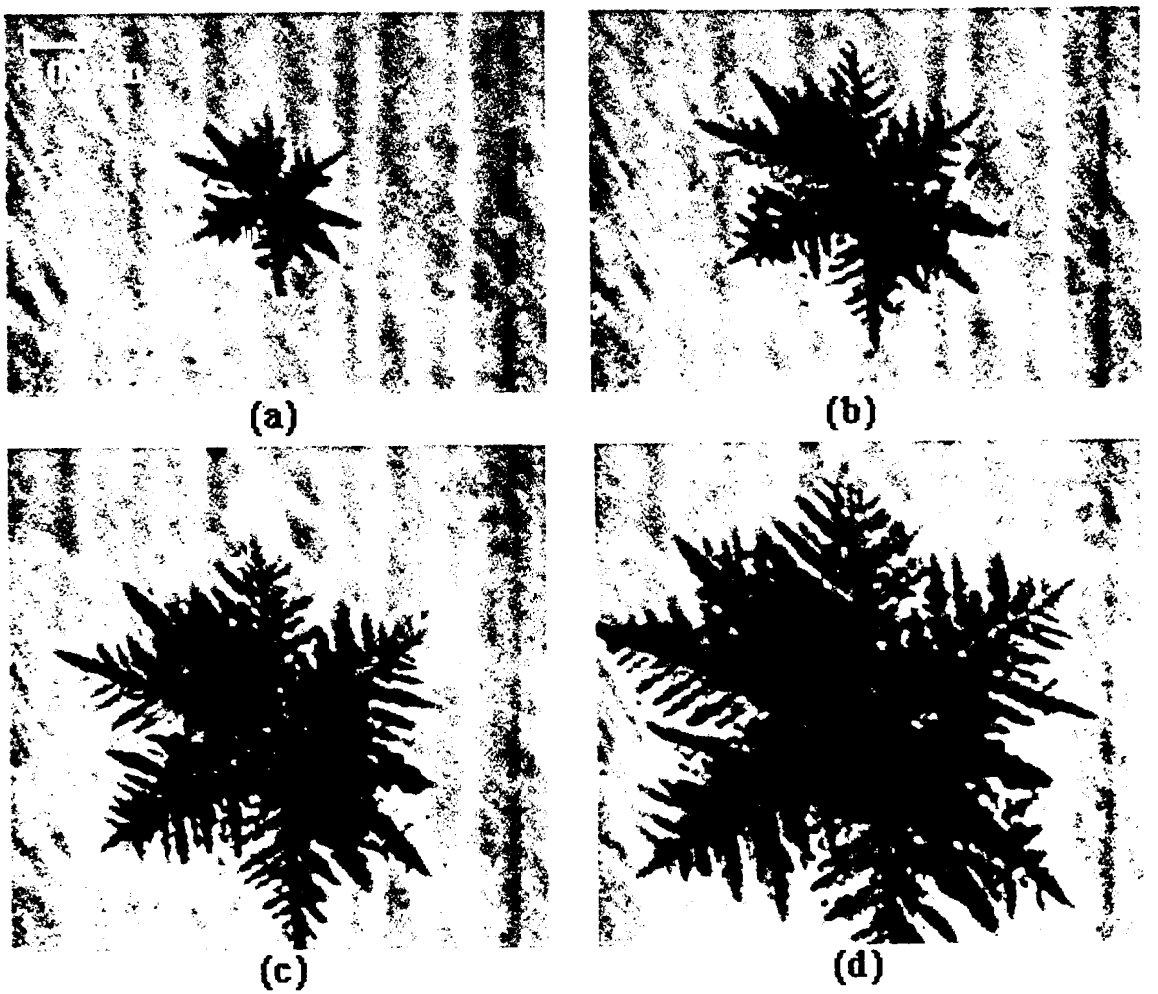

Figure 2

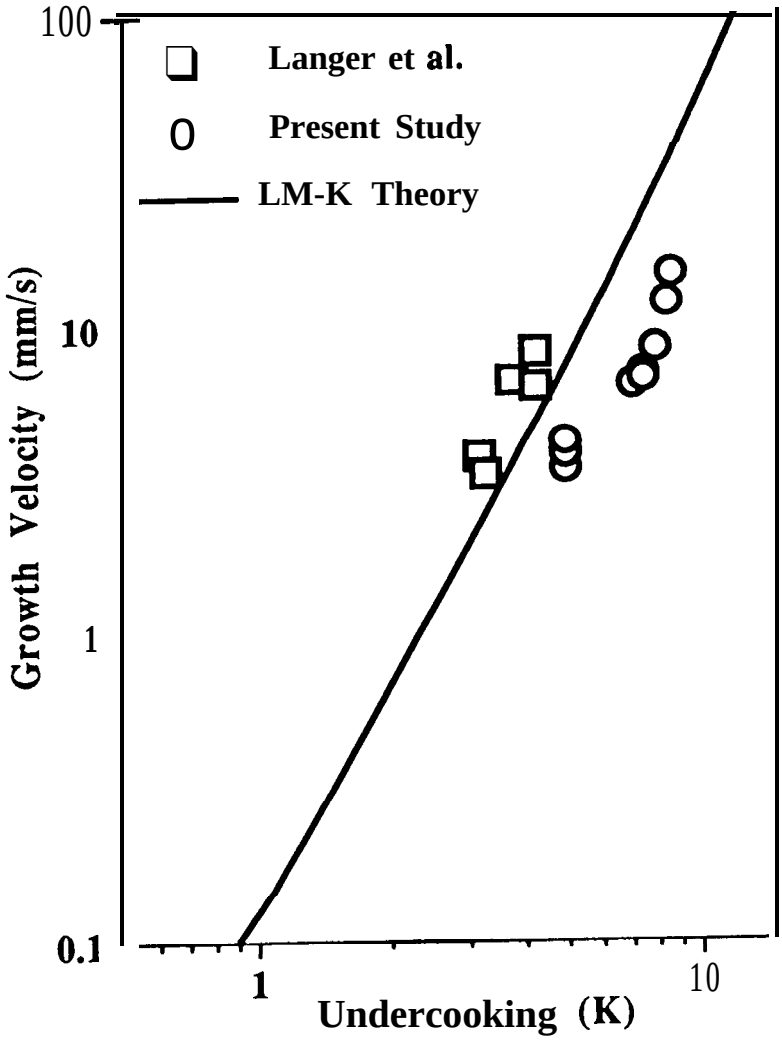
100
10
1
0.1
Growth Velocity (mm/s)
Langer et al.
Present Study
LM-K Theory
1
10
Undercooking (K)

Figure 3